PATENT SPECIFICATION

DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

Improvements in and relating to the Production of Cellulose Methyl Ethers

We, BRITISH CELANESE LIMITED, a British Company, of Celanese House, 22/23, Hanover Square, in the County of London, England, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the production of cellulose ethers containing the methyl group, for example methyl cellulose, hydroxyethyl methyl cellulose and hydroxypropyl methyl cellulose.

Methyl cellulose ethers are prepared on a commercial scale by first reacting cellulose with a caustic alkali, normally caustic soda, to form an alkali cellulose and, after a customary ageing period, reacting the alkali cellulose with methyl chloride to form the methyl cellulose. The ethers are water-soluble and in the case of the higher substituted ethers they may also be soluble in certain organic solvents such as methylene chloride-methyl alcohol mixtures. For preparing hydroxyethyl methyl cellulose the alkali cellulose is also reacted with ethylene oxide, and this reaction may be effected before or simultaneously with the methyl chloride reaction; when propylene oxide is used in the same way in place of ethylene oxide the product is a hydroxypropyl methyl cellulose.

For a number of commercial applications it is desirable that the substitution of the cellulose by the methyl groups should be as uniform as possible, and for the viscosity of solutions of the ether in water to be as close as possible to that of ether in organic solvents, for example in methylene chloride/methanol mixtures.

The present invention specifies reaction conditions for preparing the alkali cellulose and for subsequently reacting it with methyl chloride to produce methyl cellulose ethers having these desirable properties.

In the present invention the alkali cellulose is prepared with aqueous caustic soda the strength of which is closely correlated with the desired degree of substitution. The strength of the caustic soda solution is from 40 to 55 *per cent* by weight, and the temperature is so chosen that it ranges from 35° C. for the 40 *per cent* solution to 56° C. for the 55 *per cent* solution, the actual conditions used lying within the area marked out in the graph shown in the accompanying drawing. In accordance with a preferred embodiment of this invention, the conditions used are those falling within the area shown for the concentration of caustic soda ranging from 42 to 55 *per cent*.

When the alkali cellulose has been to produced it is then methylated by reaction with methyl chloride in the presence of an inert diluent in which methyl chloride is soluble. Examples of suitable diluents are ethers, for example dimethyl ether and diethyl ether. The effect of using a diluent in this way is that, in diluting the methyl chloride, heat transfer is facilitated and the reaction can be controlled as required. The diluent also functions as a vehicle in assisting the penetration of the alkali cellulose fibres by the methyl chloride.

The amount of diluent used in this way may vary between wide limits but we have obtained good results when methylating with methyl chloride containing from 45 to 90 *per cent* of its weight of dimethyl ether.

The methylation of the alkali cellulose will generally be effected after the usual ageing of the alkali cellulose. It is important that in the methylation step the elevated temperature required, usually about 72° to 78° C., should be reached as quickly as possible so as to re-

duce as far as possible any side reactions between the various compounds during the heating-up.

For producing hydroxyethyl and hydroxypropyl methyl cellulose ethers, the alkali cellulose may be additionally reacted before or during methylation with ethylene oxide or propylene oxide in accordance with known techniques.

The process of this invention has enabled us to produce improved high clarity methyl cellulose with methoxy contents ranging from 25 to 32 *per cent* which dissolve in water and, in the case of the higher substituted ethers, also in 90 *per cent* methylene chloride/10 *per cent* methanol mixtures to give solutions of comparable viscosities.

The invention is illustrated by the following examples:—

EXAMPLE 1.

320 lb. of dry cellulose sheet were immersed for 16 seconds in aqueous caustic soda of 43.6 *per cent* (w/w) strength at a temperature of 36° C. The soaked sheet was then pressed so that the ratio of wet weight to the dry weight (of cellulose) was 2.26:1.

The pressed sheet was shredded and fed to a reactor. The reactor was evacuated and then fed with 600 lb. of methyl chloride and 600 lb, of dimethyl ether. The reaction mass was raised to the reaction temperature of 77° C. in one hour and the temperature was maintained at 78° C. (±1° C.) for a further two hours.

At the end of the reaction, the dimethyl ether and excess methyl chloride were distilled to storage and the crude product was discharged. The residual alkalinity was 1.3 *per cent* (w/w) calculated as caustic soda.

The product was washed salt-free, dried and ground. Solutions made from the product were fibre-free with the following characteristics:—

Viscosity of 2 per cent aqueous solution at 20° C.: - - - - - - - - -	11,135 cps.
Clarity of 2 *per cent* aqueous solution	82 *per cent*
The methoxy content of the dry product was	26.3 *per cent.*

EXAMPLE 2.

280 lb. of dry cellulose sheet were immersed for 18 seconds in aqueous caustic soda of 52.5 *per cent* (w/w) strength at a temperature of 55° C. The soaked sheet was pressed so that the ratio of the wet weight to the dry weight (of cellulose) was 3.05:1.

The pressed sheet was shredded and fed to a reactor. After evacuation, the reactor was fed with 850 lb. of methyl chloride and 750 lb. of dimethyl ether. The reaction mass was raised to the reaction temperature of 77° C. in one hour and the temperature was maintained at 78° C. (±1° C.) for a further four hours.

At the end of this period the dimethyl ether and excess methyl chloride were distilled to storage and the crude product discharged. The residual alkalinity was 0.64 *per cent* (w/w) calculated as caustic soda.

The product was washed salt-free, dried and ground. Solutions made from the product were fibre-free with the following characteristics:—

Viscosity of a 2 *per cent* aqueous solution at 20° C.: - - - - - - - - -	1850 cps.
Clarity of a 2 *per cent* aqueous solution:	90 *per cent*
Viscosity of 2 *per cent* in 90/10 (v/v) methylene chloride/methanol at 20° C.:	1010 cps.
Clarity of 2 *per cent* in 90/10 (v/v) methylene chloride/methanol: - -	78 *per cent*

The methoxyl content of the dry product was 31.7 *per cent.*

WHAT WE CLAIM IS:—

1. A process for the production of a cellulose ether containing the methyl group which comprises reacting cellulose with an aqueous caustic soda solution having a concentration of from 40 to 55 *per cent* by weight at a temperature within the range of 35° C. to 56° C., the actual conditions of caustic soda strength and temperature lying within the area marked out in the accompanying graph, and methylating the alkali cellulose so produced by reacting it with methyl chloride in the presence of an inert diluent in which methyl chloride is soluble.

2. A process as claimed in Claim 1 wherein the conditions used for making the alkali cellulose are those falling within the area shown for the concentration of caustic soda of 42 to 55 *per cent.*

3. A process as claimed in Claim 1 or 2 wherein the inert diluent used is an ether.

4. A process as claimed in Claim 3 wherein the ether is dimethyl ether.

5. A process as claimed in Claim 4 wherein methyl chloride is used containing from 45 to 90 *per cent* of its weight of dimethyl ether.

6. A process as claimed in any of the preceding claims wherein the alkali cellulose is also reacted, before or during the methyla-

tion reaction, with ethylene oxide or propylene oxide.

7. A process as claimed in Claim 1 carried out substantially as described in either of the foregoing examples.

8. A cellulose ether containing the methyl group which has been produced by the process claimed in any of the preceding claims.

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1 SHEET *This drawing is a reproduction of the Original on a reduced scale*